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Title: Actinides and Correlated Electron Materials

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ACTINIDES AND CORRELATED ELECTRON MATERIALS

The Actinides and Correlated Electron Materials area of leadership spans Los Alamos National Laboratory competency in actinide materials research dating to the Manhattan Project as articulated in the *Integrated Plutonium Science and Research Strategy* and competency in strongly correlated electron systems dating back to at least the early 1980s.

This area of leadership focuses on the goals of **discovering, understanding, and controlling emergent electronic states and predictive performance of actinide materials.** They are quintessentially linked by the fact that the physics of actinides—and plutonium in particular—are governed by strong electronic correlations. Not only is the electronic structure of actinides dictated by fine details of electron correlations, but chemical bonding and physical structure are as well. Hence, by addressing the first goal of this leadership area we can significantly accelerate progress on the second goal. To understand such matter requires probing the intertwined spin, charge, orbital, and lattice degrees of freedom with greater precision and developing models that accurately predict the consequences of these coupled degrees of freedom, on multiple length and time scales and including acute reactivity and effects of self-irradiation phenomena in these materials.

- **Discover, understand, and control emergent electronic states:** The emphasis here is on tuning the influence of strong correlations, especially in *f*-electron systems, in diverse environments and topologies. Novel functionalities, such as unconventional superconductivity and other quantum states, frequently emerge. The existence of multiple competing or interacting degrees of freedom is common.
- **Predictive performance of actinide materials:** Meeting performance requirements from ambient to extreme conditions with prediction-driven structure-property relationships amidst aging and lifetime issues is a particular challenge with *f*-electron materials. The challenge is two-fold: 1) to forward-predict processing and aging effects on actinide performance that include a quantified uncertainty involving the identification of fundamental control variables (dose, dose rate, temperature); and 2) to develop a science-based framework for quantifying margins and uncertainties that actinide materials have on performance to gauge what we do and do not know (and how well). Understanding plutonium's 5*f* electrons plays a central role in meeting this challenge.

Cross-cutting requirements for both goals include a robust modeling and computational capability spanning multiple length and time scales to allow materials design for functionality and coupling this capability with experiments that probe and map matter across relevant length and time scales to validate predictive models and to provide new understanding.

An innovative workforce skilled in controlling emergent electronic states and in predicting performance of actinide materials is the foundation of Los Alamos National Laboratory's leadership in actinides and correlated electron materials. Shown are research technicians at work on the 40-mm gun, a key research tool for understanding the dynamic properties of plutonium.

Materials for the Future

The Los Alamos National Laboratory Materials for the Future strategy derives from our vision to support the Laboratory's national security mission drivers.

We pursue the discovery science and engineering for advanced and new materials to intentionally control functionality and predict performance relevant to ensuring the success of the Lab's missions.

To deliver on our missions, our materials strategy builds on materials science and engineering, enabling the necessary Laboratory leadership in seven key areas:

- Complex Functional Materials
- Material Resilience in Harsh Service Conditions
- Manufacturing Science
- Actinides and Correlated Electron Materials
- Integrated Nanomaterials
- Energetic Materials
- Materials Dynamics

Los Alamos Leadership in Actinides and Correlated Electron Materials

Los Alamos National Laboratory's leadership in actinides and correlated electron materials is well recognized worldwide. This recognition has grown over the decades from an institutional perspective that its workforce should be of the highest caliber, pursuing cutting-edge research, and simultaneously responding to the defense and energy needs of the country. The predictive performance of actinide materials goal is quintessential to the Laboratory's nuclear deterrence mission to ensure predictable performance of the nuclear stockpile in the absence of underground nuclear testing. However, this goal cannot be realized without the fundamental science underpinning the understanding and control of emergent electronic states.

Key Science Questions

- How do we understand—and eventually control—the emergent properties of quantum matter, which arise from complex correlations of the atomic or electronic constituents?
- What are the relevant length and time scales for controlling functionality in correlated electron materials in general, and plutonium specifically?
- What is the relevant interplay among structure, topology, correlations, pressure, temperature, magnetic field, strain rate, defects, and interfaces (including surfaces) that gives a specific response, and what is the specific role of the 5*f* electrons?
- What is beyond Fermi liquid theory as a framework for describing interacting electrons?
- Can we predict and control the time-dependent interactions of *f*-electron materials with the environment (e.g., radiation damage [including self-irradiation and external sources], interfacial chemistry, and their coupling)?

Intermediate Objectives

3-year Goals

- Invest in staff, facilities, and opportunities for interdisciplinary collaboration that span all aspects of the correlated electron problem from materials with quantum information applications to stockpile stewardship.
- Ensure Los Alamos National Laboratory is viewed as the employer of choice by increasing interaction with universities—for recruitment, retention, and pipeline development—through schools, workshops, visitor programs, etc.
- Integrate synthesis, processing, characterization, and computational capabilities to form the basis of plutonium sample, data, and modeling libraries that facilitate collaborations, data interpretation, and logistics.
- Predict, synthesize, and characterize correlated electron materials possessing novel quantum states of matter.

6-year Goals

- Global recognition of plutonium as a correlated electron problem across all institutions and disciplines (internally and externally).
- Understand phase transitions, defects, and interfaces in actinide and correlated electron materials.
- Develop experimentally validated predictive tools along with novel synthesis and measurement capabilities to enable a "materials by design" strategy of actinides and quantum materials under nonequilibrium and/or extreme conditions.
    - Extend actinide materials synthesis to isotopically pure plutonium.
- Develop and apply scale bridging approaches to enable higher fidelity treatments of materials modeling at longer time and length scales and improved descriptions of environmental interactions.

10-year End State

The ultimate goal of this area of leadership is to discover and control emergent properties for enhanced functionality spanning the atomic-, meso-, and macroscales; and connect correlation-aware predictive theory to process, age, and performance for science-based materials discovery, manufacturing, and certification. This includes the ability to predict the aging behavior of plutonium, science-based product certification and manufacturing, and Los Alamos's response to the National Quantum Initiative.

To achieve these goals, we envision the establishment of an integrated science capability through collocation, instrumentation development, strategic partnerships, and world-class facilities. This integrated capability will leverage community outreach through our institutes, user programs (Center for Integrated Nanotechnologies, National High Magnetic Field Laboratory, the Los Alamos Neutron Science Center), facilities (Plutonium Facility, Radiological Laboratory Utility Office Building, Target Fabrication Facility, Actinide Research Facility), and high-performance computing. The capability will also extend sample environments and synthesis capabilities as well as develop the highest caliber scientists who understand that correlated electrons lead to both novel quantum states of matter and the predictive performance of the actinides. Growth, characterization, measurement, computational, and modeling capabilities need to be improved for validation of experimental results. These investments will provide access to a stream of innovative ideas and position Los Alamos to be the partner and employer of choice for the leading actinide and correlated electron materials community.

For more information, please see materials.lanl.gov or send email to materials@lanl.gov.

Los Alamos NATIONAL LABORATORY

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